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AMENDED SPECIFICATION

Reprinted as amended under Section 8 of the Patents and Designs Acts, 1907 to 1932.

PATENT SPECIFICATION

Application Date: March 5, 1935. No. 6940/35.

437,236

Complete Specification Accepted: Oct. 25, 1935.

COMPLETE SPECIFICATION

A method of producing Sulphur through Reduction of Gases containing Sulphur Dioxide

I, Axel Rudolf Lindblad, a Subject of the King of Sweden, of 17 Västra Trädgårdsgatan, Stockholm, Sweden, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—

The present invention relates to an improved method of producing sulphur from gases containing sulphur dioxide and obtained by roasting or other oxidising treatment of sulphurous pyrites, arsenic pyrites, magnetic pyrites or other sulphur containing material, by reduction of the sulphur dioxide with generator gas.

It has already been proposed to recover sulphur from gases containing sulphur dioxide, by passing the said gases over a carbonaceous reducing agent at raised temperature, the reaction gases together with additional sulphur dioxide being then led into a catalyst chamber.

The essence of the present invention resides in the fact that the gases containing sulphur dioxide and a suitable quantity of oxygen are fed into a gas generator (producer) the charge of which consists of a carbonaceous material admixed or impregnated with a suitable quantity of alkali metal compound, whereupon the reducing gas mixture thus obtained is while still hot, brought together with a further regulated amount of sulphur dioxide or a gas mixture containing sulphur dioxide, and is conducted through a reaction chamber filled with a suitable catalysing material. As a result, the greater part of the supplied sulphur dioxide is reduced to elementary sulphur which is then separated in coolers and Cottrell apparatus and then collected.

It has been found that the introduction of alkali metal compounds into the generator together with the solid reducing agent has an extraordinarily favourable effect upon the entire process. Indeed, one may say that the process is practically and economically rendered possible thanks to the use of these compounds. To wit, they act as excellent catalysers upon the reduction processes included in the present method. and by introducing them at the above-mentioned part of the system together with the carbonaceous material into the generator it is possible to profit by their favourable action in all the phases of the production up to the finished sulphur. This is owing to the fact that alkali metal compounds are gasified already in the generator at the temperature prevailing in the same and then accompany, in the form of vapour or dust, the gas current through the system.

The carbonaceous material forming the charge of the gas generator may conveniently consist of coke, but other materials may also be used, for instance pit coal or charcoal, and, in some cases, it may be advantageous to feed the generator with a mixture of some of these reducing agents. Alkali metal compounds may consist of sodium compounds, which, from an economical point of view, are the cheapest, sodium carbonate, sodium sulphate or sodium sulphide having proved to be particularly adapted to the purpose in question. From a practical point of view, it is most expedient to mix or impregnate the carbonaceous material with the alkali metal compound before loading the generator with said carbonaceous material, but it is, of course, also possible to introduce the carbonaceous material and the alkaline metal compound separately.

In introducing the gas mixture containing sulphur dioxide and oxygen into the generator. which gas mixture may have been obtained in the roasting of sulphurous pyrites, arsenic pyrites or other sulphur containing material, these two gases react with the carbonaceous material with formation of sulphur and reducing gases,

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such as CO, COS and CS_2, and besides, the slight quantity of water which, in the form of moisture, is always present in the charge, and the gas mixture introduced react together to form hydrogen sulphide. The alkali metal compounds have, however, an extraordinarily catalysing effect upon these reactions, and the rapidity of reaction will be increased in a high degree through the presence of said compounds. This involves a reduction of the period of time during which the gas mixture has to be kept in contact with the carbonaceous material for obtaining a complete transformation, and this, in turn, involves that the speed of flow of the gas in the generator may be increased, thus the capacity of the same being improved to an essential degree. What this means with regard to the profitableness of the plant will be evident to everyone.

The temperature in the generator must be kept high, so that the conditions of equilibrium of those reactions which result in the formation of the above-mentioned reducing gases are as favourable as possible, a temperature comprised between 700 and 1100° C. having proved to be the most suitable one. Since the heat liberated in the process originates exclusively from the reaction of the oxygen contained in the gas mixture with the carbonaceous material, said reaction resulting in the formation of carbon monoxide, it is thus possible to set the temperature at a desired number of degrees by regulating the percentage of oxygen in the gas containing sulphur dioxide, for instance by supplying a greater or smaller quantity of air. Another possibility of regulating the temperature consists in the preheating of the gas mixture prior to its introduction into the generator, and in this manner it is possible to attain a sufficient temperature with a lower percentage of oxygen and with a higher percentage of SO_2 in the entering gas, which will, of course, be favourable from an economical point of view. Frequently it will be convenient to preheat the entering gas containing sulphur dioxide by means of gases leaving the system.

The gas generator employed may be of any construction but the best result has been obtained when the generator has been equipped with a revolving grate. Upon the reaction of the introduced gas mixture containing sulphur dioxide with the carbonaceous material there will be formed on the surface of said material an obstructing ash layer which will considerably reduce the rapidity of reaction, it being understood that the gas molecules, in entering, have to diffuse through the ash layer before reaction can take place, and, in leaving, have to diffuse through it again. All this takes a great deal of time and, therefore, it is of great importance that the obstructing ash layer be removed. This is effected in the most suitable way by using a generator fitted with movable grate, for instance a so-called revolving grate. In this case, the charge will be in permanent motion, the pieces of coal thus rubbing against each other so that the ash layer as produced will be removed according as it is formed, and free carbon surfaces will always be presented to the gas mixture.

As the reducing gas mixture coming from the generator is brought together with a regulated amount of sulphur dioxide or the gas mixture containing sulphur dioxide, usually roasting furnace gas, a reaction takes place which results in the separation of elementary sulphur in the form of vapour, essentially carbon dioxide then being formed. These reactions are catalysed by the alkali metal compound which, for the reason stated above, accompanies the reducing gas mixture coming from the generator. However, in order to cause the reduction to take place fairly quantitatively and within a reasonable period of time, it is necessary to conduct the resulting gas mixture through a reaction chamber filled with a suitable material, for instance brick, bauxite containing iron, or with other materials, especially materials containing iron, said reaction chamber being maintained at a temperature from 500 to 800° C. In this chamber the alkali metal compound contained in the gas mixture separates and so deposits on the brick contained in the chamber if such is employed, forming by reaction with silicic acid and other substances contained in the brick or other material used a catalytic coating which is highly active.

Owing to the continuous supply of alkali metal compound to the contact chamber there will, in this chamber, always exist free catalytically acting surfaces. One of the greatest disadvantages in connection with the use of a contact chamber filled with catalytic material has previously been that the surfaces of the contact substance will gradually be covered with soot, tar and dust, whereby the catalysing effect decreases and ceases in course of time. By the use of alkali metal compounds in the manner indicated above this inconvenience is removed, and free contact surfaces acting catalytically are always presented to the gas mixture.

In order further to illustrate the principle of the present invention there will be given an example which gives a good result of the manner in which the method may be carried out. This example should, however, only be regarded as an example of various embodiments of the invention, which embodiments may be used in the application of the method in practice, and thus does not involve a limitation of the invention to this embodiment only.

The percentage of sulphur dioxide in those gases which are used in the present process, for instance roasting furnace gases, may be of any order of magnitude, but it is advantageous for them to contain, on their introduction into the gas generator, about 4 to 5 per cent. SO_2 and an amount of oxygen of about 11 per cent. by volume O_2 (free, i.e. not chemically combined oxygen). The roasting of sulphurous pyrites or other sulphur containing material may, of course, be carried on in such a manner that a gas having this composition will be obtained, it is true, but generally it will be more suitable, with a view to later phases of the manufacture, to try to produce a roasting furnace gas having as high a percentage of SO_2 as possible, for instance 9 to 10 per cent. SO_2 and an amount of oxygen of 4 to 5 per cent. O_2, whereupon this gas is diluted with a regulated amount of air, so that the resulting gas mixture will contain about 11 per cent. O_2. This gas, prior to its introduction into the gas generator, is conducted through a preheater in which it is heated from 100 to 300° C. by means of hot exhaust gases leaving the system. By supplying gas having this temperature and this percentage of oxygen to the gas generator there will be obtained in the generator a temperature favourable to the process and amounting in the present case, to about 1000° C.

The carbonaceous material forming the charge of the gas generator provided with revolving grate conveniently consists of coke in which has been incorporated about 1 percentage by weight of sodium carbonate. This amount of sodium carbonate has proved to be satisfactory to bring about a catalysing effect that is quite sufficient for the process.

The hot gas mixture containing sulphur dioxide and being rich in oxygen, after having been introduced into the generator, reacts, while being influenced by the soda catalysing the reaction, with the reducing agent with formation of sulphur, carbon monoxide, carbon oxy-sulphide and carbon disulphide, the sodium carbonate being gasified to a larger or smaller extent and accompanying, in the form of vapour or dust, the reducing gases leaving the generator, on their way through the system.

The reducing gases escaping from the charge of the generator are, while being still hot, possibly in the upper part of the generator or immediately outside the same, brought together with a further amount of gas containing sulphur dioxide, said amount of gas being regulated according to circumstances. A roasting furnace gas having the composition indicated above, i.e. containing about 9 to 10 per cent. SO_2 and about 4 to 5 per cent. O_2 is particularly adapted to this purpose. As a result, a reaction takes place which is catalysed by the sodium compound present, elementary sulphur then being formed. In order to cause the reaction to take place fairly quantitatively it is, however, necessary to conduct the gas mixture as produced to a contact chamber filled with a suitable material, preferably a material containing iron. Said material may consist of ordinary brick or, what is still more preferable, of bauxite, and the temperature in the contact chamber should amount to 500 to 800° C. The sodium compound which has accompanied the gas mixture separates in said chamber and deposits on the surfaces of the contact material, while forming a catalytic coating which is extraordinarily active and, due to the continuous supply of sodium compound, will be incessantly renewed so that free surfaces acting catalytically are always presented to the gas mixture. In this contact chamber, the reaction between the reducing gases and the sulphur dioxide is carried on to an end, whereupon the sulphur formed is separated in coolers and Cottrell apparatus.

Thus, the alkali, for instance sodium carbonate, introduced into the gas generator is utilised in two different phases or moments during production, in the gas generator as well as in the catalyser chamber, to which it is carried, as mentioned above, in gaseous state or in the form of powder or dust.

To summarise, it may be said that the inventor, by supplying alkali metal compounds to the gas generator together with the charge of the same and by effecting the reduction process in the manner hereinbefore indicated, has succeeded in introducing, in a single procedure, an extraordinarily active catalyser, the action of which may be utilised throughout the system. Of course, the same amount of alkali metal compound catalyses, on the one hand, the reaction of the gas mixture containing sulphur dioxide and oxygen with the carbonaceous material, and, on the other hand, the reduction of the supplied gas containing sulphur dioxide and usually consisting of roasting furnace gas, which reduction is effected by means of the reducing gases as formed. The action of the alkali metal compounds thus extends from the beginning of the process to the end thereof, and one can speak of a complete utilisation of the same.

In my Patent Specification No. 428,979 I have described and claimed a similar method of producing sulphur from gases containing SO_2 wherein the gases prior to their reduction with carbonaceous material are cooled, treated in an electrostatic purifier and then reheated. The reduction may be effected by means of a carbonaceous material in the presence of an alkali metal compound, the resulting gas after admixture with a further quantity of gas containing SO_2 being brought into contact with a catalyst.

Having now particularly described and ascertained the nature of my said invention and in what manner the same is to be performed, I declare that I make no claim herein to what is claimed in the specification of my Patent No. 428,979 but subject to this disclaimer what I claim is:—

1. In the production of sulphur from gases containing SO_2 and obtained by the roasting or other treatment of sulphurous pyrites, arsenic pyrites, magnetic pyrites or other sulphuriferous materials the method according to which gas mixture containing sulphur dioxide and oxygen is, in a gas generator, brought together with carbon or carbonaceous material which has been mixed or impregnated with alkali metal compound whereupon the resulting reducing gas mixture together with alkali metal compound included in the same in the form of vapour or dust is, in a hot state, brought together with a further amount, suitably regulated for the purpose, of sulphur dioxide or a gas mixture containing sulphur dioxide and is conducted through a reaction chamber (catalyser chamber) containing a material which acts catalytically.

2. A method as claimed in Claim 1, characterised in that the temperature in the gas generator is regulated by varying the percentage of oxygen in the supplied gas mixture.

3. A method as claimed in Claim 1, characterised in that the gas mixture containing sulphur dioxide and oxygen is preheated prior to its introduction into the gas generator.

4. A method as claimed in Claim 1, characterised in that alkali metal compound is introduced separately into the gas generator and is permitted to accompany the gas mixture, in the form of vapour or dust, into the catalyser chamber.

5. A method as claimed in Claim 1, characterised in that the carbonaceous material is impregnated with a sodium compound, for instance sodium carbonate, sodium sulphate or sodium sulphide.

6. A method as claimed in Claim 1, characterised in that the gas generator employed is fitted with a movable grate, for instance a revolving grate arranged in a manner known per se.

7. The improved method of producing sulphur from gases containing sulphur dioxide substantially as described.

Dated this 5th day of March, 1935.

MARKS & CLERK.

Leamington Spa: Printed for His Majesty's Stationery Office, by the Courier Press.—1936.